

# Kinetics of Deposition in the Diffusion-Controlled Limit

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We study the adsorption of particles diffusing in a half-space bounded by the substrate and irreversibly sticking to the substrate upon contacts. We show that when absorbing particles are planar disks diffusing in the three-dimensional half-space, the coverage approaches its saturated ‘jamming’ value as  $t^{-1}$  in the large time limit [generally as  $t^{-1/(d-1)}$  for the adsorption on the  $d$ -dimensional substrate when  $d > 1$  and as  $e^{-t/\ln(t)}$  when  $d = 1$ ]. We also analyze the asymptotic behavior when particles are planar aligned squares and when they are spherical.

## I. INTRODUCTION

The deposition of suspended particles onto a clean substrate is an important process in physics, chemistry, biology, and technology [1–3]. Adhesion of colloidal particles and proteins on substrates are just a few examples of numerous applications. A comprehensive description of the deposition process is very challenging as it requires the understanding of the evolution of a strongly interacting infinite-particle system. Indeed, suspended particles diffuse and directly interact with each other through exclusion. Additionally, the motion of suspended particles causes long-ranged hydrodynamic inter-particle interactions. Further, the process of attaching to the substrate is also very complicated, the particles may rotate, the shape of the particles plays an important role, etc. Even when the volume fraction occupied by suspended particles is very small and hence the inter-particle interactions in the solution are ignored, very little is known theoretically. It is therefore customary to maximally simplify the problem. Following this tradition we focus on dilute systems, we ignore rotations and we consider particles of a few simple shapes. Most studies also ignore diffusion. In contrast, our major goal is to probe the influence of diffusion of suspended particles. We therefore focus on the diffusion-controlled limit in which adhesion occurs instantaneously and diffusion plays the dominating role.

The full treatment of the diffusion-controlled deposition problem is beyond the reach of analytical approaches, so we shall employ heuristic arguments. One particular situation amenable to heuristic treatment is when the particles are planar disks diffusing in a half-space above the flat substrate. Disks are assumed to remain parallel to the substrate. Whenever a disk touches the substrate, it irreversibly adheres to it. The overlapping of disks on the substrate is forbidden. The substrate eventually reaches a jammed state that cannot accommodate additional disks. What fraction of the substrate is covered in the jammed state? What is the temporal evolution in the vicinity of the jammed coverage? Are there correlations in particle positions in the jammed state?

Some progress in answering such questions has been reached in the setting which totally ignores the diffusion of the particles in the solution. This framework is known as the random sequential adsorption (RSA). The RSA model postulates that the deposition events

are random: If the new particle does not overlap with already deposited ones, it sticks to the substrate; otherwise, the deposition event is discarded. The RSA model has been introduced long time ago [4–6] and it is fairly well-understood (see [7–10] for a review), although analytical solutions have been established only in the case of the one-dimensional substrate (when disks become segments). For the RSA of disks the basic properties of the jammed state like the jamming coverage  $\rho_{\text{jam}}$  are unknown. The asymptotic approach to the jamming coverage is known [11–13], namely  $\rho_{\text{jam}} - \rho(t) \sim t^{-1/2}$  for the RSA of disks. This result admits a generalization to arbitrary dimension [12, 13]:

$$\rho_{\text{jam}} - \rho(t) \sim t^{-\sigma} \quad (1)$$

for  $t \gg 1$  with jamming exponent  $\sigma = 1/d$  for the RSA of disks onto the  $d$ -dimensional substrate.

The RSA model is a single-particle process with memory. Indeed, in each deposition event one particle is involved and the positions of already deposited particles are required. The diffusion-controlled deposition process studied below is a truly infinite-particle system. In the next Sect. II we show that for the adsorption of diffusing disks the asymptotic approach to the jamming coverage is also algebraic, and we determine the jamming exponent. We then analyze the diffusion-controlled deposition process when the diffusing particles are aligned planar squares (Sect. III) or spheres (Sect. IV). We conclude with a short discussion (Sec. VI).

## II. ADSORPTION OF DIFFUSING DISKS

We emphasize again the basic assumptions of the diffusion-controlled adsorption of discs:

1. Disks freely diffuse (no interactions) in the solution and they remain parallel to the substrate.
2. Adsorbed disks do not overlap, do not desorb from the substrate, and do not diffuse on the substrate.

Even for planar particles one can consider the general situation when they do not remain parallel to the substrate. Furthermore, in the case of planar particles which remain parallel to the substrate, the analysis becomes more challenging when particles are different from

disks. Below we shall comment on these more complicated systems, but now we limit ourselves by diffusing planar disks irreversibly sticking to the substrate.

In this section we argue that

$$\rho_{\text{jam}} - \rho(t) \sim t^{-1} \quad \text{when } d = 2 \quad (2)$$

Generally the jamming exponent underlying the adsorption of diffusing disks is given by

$$\sigma = \frac{1}{d-1} \quad (3)$$

when  $d > 1$ . The jamming exponent (3) becomes infinite if  $d = 1$ . This means that the algebraic approach (1) is no longer valid. (The disks are segments when  $d = 1$ , they remain parallel to one-dimensional substrate, diffuse in the two-dimensional half-space and adhere to the substrate upon touching it.) We shall argue that for  $d = 1$  the asymptotic approach to the jamming coverage is just slightly slower than pure exponential:

$$\rho_{\text{jam}} - \rho(t) \sim \exp(-Ct/\ln t) \quad (4)$$

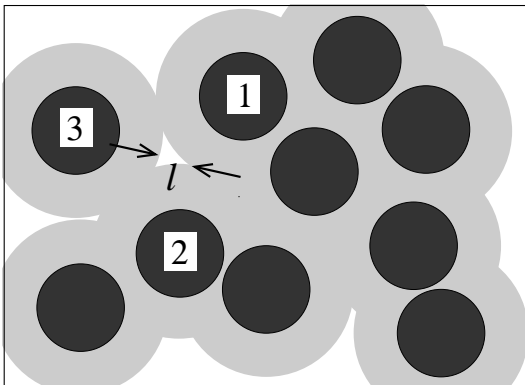


FIG. 1: Illustration of a target zone (white) between disks 1, 2, and 3. Exclusion zones (shaded) and the adsorbed disks (dark) are also shown. In the long-time limit target zones are mostly triangular and they are far away from each other. (Adapted from Ref. [10].)

Let us first establish (2). Around each adsorbed disk there is an exclusion zone (its radius is twice that of the disk, Fig. 1). When a new disk from the bulk adsorbs, its center touches the substrate at a target zone. As the substrate approaches jamming, the target zones become small and almost all of them acquire triangular shapes (more precisely, they have arcs-shaped sides whose radius is twice that of the disks, see Fig. 1). The separation between target zones grow indefinitely and in a jammed configuration there are no target zones. Hence in the

long time regime we can ignore the interaction between target zones. This greatly simplifies the determination of the asymptotic behavior of the coverage and it was the chief idea of Pomeau [12] and Swendsen [13] in the case of the RSA model that led to (1) with  $\sigma = 1/d$ . This remains valid in our case, although the elimination of the target zones proceeds in a different manner.

Denote by  $c(\ell, t)$  the density of target zones of linear size  $\ell$ . In the long time limit almost all target zones are very small,  $\ell \ll R$  where  $R$  is the radius of disks. The density  $c(\ell, t)$  decays according to the rate equation

$$\frac{dc(\ell, t)}{dt} \simeq -A_2 n_\infty D \ell c(\ell, t) \quad (5)$$

where  $D$  is the diffusion coefficient,  $n_\infty$  is the density of the disks far away from the target, and  $A_2$  is a numerical factor. Accepting (5) we get

$$\begin{aligned} \rho_{\text{jam}} - \rho(t) &\sim \int_0^R \frac{d\ell}{R} c(\ell, t) \\ &\sim \int_0^R \frac{d\ell}{R} e^{-A_2 n_\infty D \ell t} \sim (n_\infty R D t)^{-1} \end{aligned} \quad (6)$$

in agreement with the announced decay law (2).

For the  $d$ -dimensional substrate with  $d > 1$ , an analog of (5) is

$$\frac{dc}{dt} \simeq -A_d n_\infty D \ell^{d-1} c \quad (7)$$

from which we deduce

$$\rho_{\text{jam}} - \rho(t) \sim R^{-1} (n_\infty D t)^{-\frac{1}{d-1}} \quad (8)$$

To understand (5) and (7), consider a small isolated triangular target zone. (We emphasize again that the separation between target zones diverges and their sizes are small,  $\ell \ll R$ , when  $t \rightarrow \infty$ .) For dilute suspensions we can neglect the exclusion volume interaction between the disks. (In Sect. V we outline extensions to dense suspensions.) Thus we can treat the centers of the disks as non-interacting Brownian point particles. The triangular zone is eliminated when a point particle touches it. We can disregard the reflection boundary condition on the substrate by considering a ‘two-sided’ problem, namely a planar  $d$ -dimensional target zone at  $z = 0$  in  $\mathbb{R}^{d+1}$ .

The elimination of any target is described by reaction rate theory [10, 14] which expresses the decay rate through the diffusion coefficient  $D$ , the density  $n_\infty$  of the disks far away from the target, and the capacitance  $C$  of the target. (A connection with electrostatics goes back to Berg and Purcell [15], see [10] review.) When  $d = 2$ , so the ambient space is three-dimensional, the decay rate is  $4\pi n_\infty D C$ . Taking into account that our targets are planar and adsorption is possible only from above we conclude that the proper decay rate in our case is  $2\pi n_\infty D C$ . Reaction rate theory is applicable when  $d > 1$  and the general prediction for the decay rate of a planar target

is  $\frac{1}{2}\Omega_{d+1}n_\infty DC$ , where  $\Omega_{d+1} = 2\pi^{(d+1)/2}/\Gamma[(d+1)/2]$  is the area of the unit sphere  $\mathbb{S}^d$  in  $\mathbb{R}^{d+1}$ . The capacitance scales as  $\ell^{d-1}$  where  $\ell$  is the characteristic size of the target zone (when all dimensions of the target zone are comparable) [16]. Thus the decay rate is proportional to  $n_\infty D\ell^{d-1}$  explaining (5) and (7).

The capacitance of the triangular target zone depends on its shape. For the disk of radius  $\ell$ , for instance, the capacitance is  $C = 2\ell/\pi$  [16], so the decay rate would be equal to  $4n_\infty D\ell$  if the target zone was the disk. Generally for triangular target zones the decay rate is  $A_2 n_\infty D\ell$  with a numerical factor  $A_2$  of order unity depending on the details of the shape of the zone.

The reaction rate theory is slightly different [10] in the case of the two-dimensional ambient space ( $d = 1$ ). The density  $c(\ell, t)$  of the target zones of length  $\ell$  decays almost exponentially,

$$c(\ell, t) \sim \exp\left[-\frac{2\pi n_\infty Dt}{\ln(Dt/\ell^2)}\right], \quad (9)$$

from which a more precise version of (4),

$$\rho_{\text{jam}} - \rho(t) \sim \exp\left[-\frac{2\pi n_\infty Dt}{\ln(Dt/R^2)}\right] \quad \text{when } d = 1, \quad (10)$$

is obtained.

### III. ADSORPTION OF DIFFUSING SQUARES

To analyze the influence of the shape of the diffusing planar objects on the dynamics of the deposition process let us consider the deposition of aligned squares. In the realm of the RSA this problem has been extensively studied, see e.g. [6, 13, 17–20] and reviews [7–9].

We assume that the squares are identical, say of the size  $R \times R$ , and their attachment to the substrate is aligned with  $x$  and  $y$  axes. We treat the deposition process heuristically using the same approach as in the previous section. The target zones are now rectangles. Long-lived  $\ell \times L$  rectangles have sizes smaller than the linear size of the square:  $\ell < R$  and  $L < R$ . Crucially, long-lived rectangles tend to have a very large aspect ratio. The capacitance of an  $\ell \times L$  rectangle with  $L \gg \ell$  is  $C \sim L/\ln(2L/\ell)$ . This formula gives a correct qualitative behavior even for  $L \sim \ell$ . More accurate formulas were already known to Maxwell [21] and the work on this issue is continuing (see e.g. [22] and references therein), but the above is sufficient for heuristic reasoning.

The reaction rate theory tells us that the flux to the target zone scales as  $n_\infty DC$ , so

$$c(\ell, L; t) \sim \exp\left[-A_2 n_\infty Dt \frac{L}{\ln(2L/\ell)}\right] \quad (11)$$

The coverage saturates according to

$$\begin{aligned} \rho_{\text{jam}} - \rho(t) &\sim \int_0^R \frac{d\ell}{R} \int_\ell^R \frac{L}{R} c(\ell, L; t) \\ &\sim \int_0^R \frac{d\ell}{R} \frac{\ln(2R/\ell)}{n_\infty R Dt} e^{-A_2 n_\infty D\ell t} \\ &\sim \frac{\ln(n_\infty R Dt)}{(n_\infty R Dt)^2} \end{aligned} \quad (12)$$

This decay law is faster than the  $t^{-1} \ln(t)$  decay [13, 20] characterizing the asymptotic approach to the jamming coverage in the realm of the RSA of aligned squares.

### IV. ADSORPTION OF DIFFUSING SPHERES

Diffusing particles can have various shapes. The case of diffusing spheres (Fig. 2) is especially interesting, a suspension of latex spheres is an obvious example. The adsorption of diffusing spheres has been modeled in Ref. [23] in the most relevant case of  $d = 2$ . The  $t^{-2/3}$  approach to the jammed state was predicted. This approach is faster than the  $t^{-1/2}$  approach characterizing the RSA of spheres, but slower than the  $t^{-1}$  decay law (2) describing the deposition of disks in the diffusion-controlled limit. In this section we re-derive the  $t^{-2/3}$  behavior and show that generally for  $d > 1$  the jamming exponent is

$$\sigma = \frac{2}{2d-1} \quad (13)$$

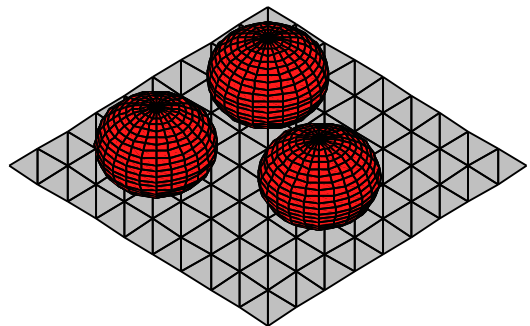


FIG. 2: Spheres attached to the substrate are more than the disks, they affect diffusing spheres above the substrate causing them to go through the narrowing channels.

In one dimension, the exponent  $\sigma = 2$  predicted by (13) is correct, but there is also an additional logarithmic correction. We will argue that the decay law is

$$\rho_{\text{jam}} - \rho(t) \sim \left(\frac{\ln t}{t}\right)^2 \quad \text{when } d = 1 \quad (14)$$

At first sight, there seems to be no difference between adsorption of planar disks and spheres. In the RSA framework these two adsorption processes are *identical*. Adding diffusion changes the situation (Figs. 2 and 3). In the case of spheres we can still focus our attention on the deposition of the point particles, the centers of diffusing spheres, into target zones. However to reach the zone, the particle must go through the channel and reach its bottleneck.

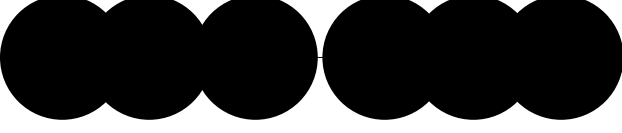


FIG. 3: Two-dimensional illustration when spheres become (vertical) disks. The radius of each disk is twice the radius of the sphere, so it represents the excluded region for the centers of diffusing spheres. The center of the new sphere must go through the channel between two central disks and it must reach the bottleneck (the horizontal segment).

Reaction rate theory implies that to compute the flux it suffices to replace the diffusion equation by the (stationary) Laplace equation

$$\nabla^2 n = 0 \quad (15)$$

This replacement is justified in the case when the ambient space is three-dimensional (more generally when  $d > 1$ ). Far away from the target

$$n = n_\infty \quad (16)$$

The density must also satisfy the absorbing boundary condition on the target (the bottleneck in Fig. 3)

$$n|_{\text{inside target}} = 0 \quad (17)$$

and the reflecting boundary condition on the boundary of the excluded region. This latter boundary is complicated (see Fig. 3) and random as it depends on the history of the deposition process. When the target zone is very small, however, i.e.  $\ell \ll R$ , we can significantly simplify the problem. The passage through the very narrow channel is governed by an ‘entropic’ barrier which provides the dominant contribution to the decay rate. (Problems with entropic barriers are analyzed in numerous studies, see e.g. [24–27] and references therein.) In the channel depicted in Fig. 3, the three-dimensional Laplace equation (15) can be replaced by the quasi-one-dimensional Laplace equation

$$\frac{d}{dy} \left[ A(y) \frac{dn}{dy} \right] = 0 \quad (18)$$

where  $A(y)$  is the cross-section area of the channel at the height  $y$  above the bottleneck.

In the case of the one-dimensional substrate represented in Fig. 3, if  $\ell$  is the width of the bottleneck then the width of the channel is  $A(y) = \ell + \frac{y^2}{R}$  when  $y \ll R$ . In the case of the two-dimensional substrate the cross-section area scales as  $A(y) \sim \left( \ell + \frac{y^2}{R} \right)^2$ . Using this estimate and integrating (18) we get

$$\left( \ell + \frac{y^2}{R} \right)^2 \frac{dn}{dy} = F \quad (19)$$

Integrating one more time we obtain

$$n = F \sqrt{\frac{R}{\ell^3}} \int_0^{y/\sqrt{R\ell}} \frac{d\eta}{(1 + \eta^2)^2} \quad (20)$$

Using the boundary condition (16) we fix the constant

$$F \sim n_\infty \sqrt{\frac{\ell^3}{R}} \quad (21)$$

The flux is given by  $DF$ . Therefore the density  $c(\ell, t)$  of target zones of linear size  $\ell$  decays according to

$$\frac{dc(\ell, t)}{dt} \sim -Dn_\infty \sqrt{\frac{\ell^3}{R}} c(\ell, t) \quad (22)$$

from which we deduce

$$\rho_{\text{jam}} - \rho(t) \sim (n_\infty R D t)^{-2/3} \quad (23)$$

Generally when  $d > 1$  we use the stationary Eq. (18) with  $A(y) \sim \left( \ell + \frac{y^2}{R} \right)^d$  and arrive at

$$\frac{dc(\ell, t)}{dt} \sim -Dn_\infty \sqrt{\frac{\ell^{2d-1}}{R}} c(\ell, t)$$

which leads to the announced exponent (13). More precisely, the coverage saturates according to

$$\rho_{\text{jam}} - \rho(t) \sim (n_\infty R^{d-1} D t)^{-\frac{2}{2d-1}} \quad (24)$$

The case of the one-dimensional substrate is more subtle. Strictly speaking, one cannot employ the stationary framework. An asymptotically correct results can be established using a simple trick: One formally solves Eq. (18) to yield

$$n = F \sqrt{\frac{R}{\ell}} \int_0^{y/\sqrt{R\ell}} \frac{d\eta}{1 + \eta^2} \quad (25)$$

and one matches this solution to  $n_\infty / \ln(R^2 D t)$  instead of  $n_\infty$ . We thus find

$$\ln c(\ell, t) \sim -\frac{n_\infty D t}{\ln(R^2 D t)} \sqrt{\frac{\ell}{R}}$$

which leads to Eq. (14), more precisely to

$$\rho_{\text{jam}} - \rho(t) \sim \left[ \frac{\ln(R^2 D t)}{n_\infty D t} \right]^2 \quad (26)$$

	RSA		Diffusion-controlled		
substrate dimension	disks or spheres	aligned squares	disks	aligned squares	diffusing spheres
$d = 2$	$t^{-1/2}$	$t^{-1} \ln t$	$t^{-1}$	$t^{-2} \ln t$	$t^{-2/3}$
$d = 1$	$t^{-1}$	$t^{-1}$	$e^{-t/\ln t}$	$e^{-t/\ln t}$	$t^{-2}(\ln t)^2$

TABLE I: The asymptotic approach to the jamming coverage for the RSA and for the diffusion-controlled deposition process. In one dimension, the aligned squares and disks are just segments, so the behavior is the same.

## V. DENSE SUSPENSIONS

In this section we show how in principle one can take into account the exclusion volume interaction. A general approach allowing to treat the survival probability of a trap in diffusive *lattice* gases has been recently developed [28]. Some lattice gas models, e.g. so-called symmetric exclusion process, have been analyzed in details. Similar behaviors are expected to hold in the present case of *continuous* gases of suspended particles. For  $d > 1$ , the jamming exponent is not affected by exclusion volume interaction. For  $d = 1$ , the time dependence also remains the same. The new feature is the non-trivial dependence on the volume fraction  $n_\infty R^d$  occupied by the particles. For instance, in the case of adsorption onto the plane

$$\rho_{\text{jam}} - \rho(t) \sim \frac{\Phi(n_\infty R^3)}{n_\infty R D t} \quad (27)$$

When  $n_\infty R^3 \ll 1$ , the exclusion volume interaction can be ignored:  $\Phi(\nu) \sim 1$  when  $\nu \rightarrow 0$  and we recover (6). To compute the function  $\Phi(\nu)$  in the case of dense suspensions seems impossible.

The formal scheme of the computation [28] relies on a macroscopic fluctuation theory (see [29] for a review). In the long time limit it suffices to consider a single target zone. The logarithm of the probability that it remains uncovered at time  $t$  is asymptotically

$$-\ln S \simeq \frac{1}{2} t \int d\mathbf{r} \frac{D^2(n)}{\sigma(n)} (\nabla n)^2 \quad (28)$$

when  $d > 1$ . The integral in Eq. (28) is taken over the  $(d+1)$ -dimensional ambient space outside the target zone. The integrand in (28) contains two transport coefficients, the diffusion coefficient  $D(n)$  and the mobility  $\sigma(n)$ . The density  $n(\mathbf{r})$  is determined by the solution of a *stationary* partial differential equation

$$\nabla^2 n + \Psi(n)(\nabla n)^2 = 0, \quad \Psi(n) = \frac{d}{dn} \left[ \ln \frac{D(n)}{\sqrt{\sigma(n)}} \right] \quad (29)$$

subject to (16)–(17) and the reflecting boundary condition on the boundary of the excluded region in the case of the suspension of spheres. The case of planar disks

is again particularly simple as we do not need the latter boundary condition, we can instead analyze (29) in  $\mathbb{R}^{d+1}$ .

It is usually impossible to solve the non-linear partial differential equation (29) subject to the boundary conditions (16)–(17). For a few simple lattice gas models explicit analytical solutions were found in [28], and also in Ref. [30] for bounded domains. Some formal analytical solutions were also established [28, 30], but one should keep in mind that for almost all interacting gases the transport coefficients  $D(n)$  and  $\sigma(n)$  are unknown. For simple gases interacting only through exclusion the diffusion coefficient is density-independent, but the mobility is still very hard to determine—one needs to know the free energy, and in two and higher dimensions it is essentially impossible for any gas with exclusion interactions.

## VI. DISCUSSION

Table I represents the asymptotic behaviors for the particles of three different shapes (planar disks, planar aligned squares and spheres) for the diffusion-controlled and random sequential adsorption processes. In all these cases the approach to the jamming coverage for the diffusion-controlled deposition processes is significantly faster than for the RSA.

The diffusion-controlled deposition of other objects (e.g. planar ellipses) is an interesting avenue for further research. The shape and the orientational freedom of the depositing objects may affect the asymptotic behaviors. For instance, one would like to explore the diffusion-controlled deposition of planar ellipses (planar squares) in the case of the isotropic deposition, i.e., assuming that the ellipses (squares) undergo both translational and rotational diffusion). In the realm of the RSA the asymptotic behaviors depend on symmetries of the objects and the orientational freedom [9, 17–20], but in the isotropic case the universal  $t^{-1/3}$  behaviors emerges for squares (and generally for rectangles) and for ellipses with arbitrary non-zero eccentricity [17–19].

In the deposition processes studied above the substrate eventually gets clogged. This is not necessarily the case when the depositing objects have no width. For the RSA of needles, for instance, the deposition process never ends although the growth slows down, namely the density of the needles per unit area increases as  $t^{\sqrt{2}-1}$  [31], see also Refs. [32, 33] for further results and analyses of related fragmentation problems. Therefore the RSA of needles is characterized by an irrational exponent. The diffusion-controlled deposition of needles is an intriguing open problem.

I am grateful to Julian Talbot for interesting discussions. This research was supported by grant No. 2012145 from the BSF.

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